International Journal of Mechanical and Production Engineering Research and Development (IJMPERD)
ISSN(P): 2249-6890; ISSN(E): 2249-8001
Vol. 9, Issue 4, Aug 2019, 1087-1096

POSSIBILITY OF PRODUCING SINTERED FINE POROUS GRANULATED CERAMIC FILLER USING ASH OF THERMAL POWER STATIONS IN COMBINATION WITH CLAY ROCKS

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ABSTRACT

In localities where rocks for production of coarse and fine fillers are unavailable, it becomes necessary to develop innovative artificial materials based on processing of local nonconventional natural and manmade feed stocks. This work presents experimental studies devoted to development of sintered fine porous granulated ceramic filler based on ash of thermal power station in combination with clay rocks. Clay–ash ceramic composition has been studied. Ash content in this composition was from 10 to 30%. Granules of the ceramic composition were sintered in rotary kiln at 1,000°C. Main features of physico-mechanical properties of samples were studied as a function of ash content up to 30%. With addition of ash of Ekibastuz Power Station in amount from 10%to 30%, the strength of granules increases. Thus, at annealing temperature of 1,000°C, the strength increases to 12.4 MPa. Here with, the average density decreases and is from 1,250 kg/m^{3}to 910 kg/m^3. The obtained experimental data also evidence intensification of sintering and crystallization in ceramic pastes containing ash of Ekibastuz Power Station. Samples without ash addition are characterized by dense sintered structure with insignificant micropores. Granules with ash addition to 30%are characterized by developed sintered porous structure. It has been established that keramdor materials based on the considered composition are characterized by strong microporous structure after annealing. In addition, ash of Ekibastuz Power Station in the ceramic paste exerts favorable influence on heat conducting properties of sintered microporous granulated ceramic filler, which makes it possible to use them as material, increasing energy efficiency of buildings and structures. The use of ash in ceramic paste for production of granulated sintered microporous material allows improving environmental protection, partially.

***KEYWORDS:** Ash, Ceramic Composition, Granulated Fine Porous Ceramic Filler, Burning, Rotary Kiln & Crystalline Phase*

Received: May 20, 2018; **Accepted:** Jun 10, 2019; **Published:** Jul 16, 2019; **Paper Id.:** IJMPERDAUG2019112

Original Article

INTRODUCTION

The rates of residential, industrial, road and agricultural construction in Kazakhstan are increasing continuously. In addition, new oil and gas deposits are developed, new oil and gas pipelines are constructed.

In order to successfully fulfill these global state targets, it is required to apply innovative scientific approaches to production of efficient building materials. The major portion in construction is that of coarse and fine fillers for production of concretes, reinforced concrete structures, bitumen concretes, ballast of road structure,

as well as structural elements upon construction of special purpose facilities (bridges, tunnels, ducts, etc.).

In general, the share of coarse and fine fillers in the aforementioned items and structures, including that of automobile and rail roads, is about 65-85%. However, deposits of rocks suitable for production of coarse and fine fillers in Kazakhstan are distributed non-uniformly.

As a consequence, coarse and fine fillers are delivered to remote regions by railway and automobiles. Transportation of high amounts of these materials significantly increases their prime cost. For instance, gravel produced in Aktyubinsk is sold at the price of 1,000-1,500 tenge per 1 t. Transportation of this gravel to Uralsk (West Kazakhstan oblast, 550-600 km) increases the price to 5,500-6,500 tenge per 1 t, that is, the price increases more than by 5 times. Such situation leads to general increase in prime costs of all erected facilities and road construction.

In the areas where rocks for production of coarse and fine fillers are unavailable, it is necessary to develop innovative artificial materials based on wastes, nonconventional local feedstocks of natural and manmade origins.

Therefore, the issues of improvement of production of artificial fillers are urgent with regard to involvement of new feedstocks, improvement of process output in terms of energy efficiency and resource savings.

One of the promising materials providing partial or complete replacement of natural coarse fillers is ceramic road construction material (keramdor). This is sintered (thermally treated) granulated material with various particle sizes produced by processing of local clay rocks.

Numerous versatile studies in this field are available (Galuzin, 1969; Trusov, 1972; Beskrovnyi etal, 1970). All these studies are devoted to development of production of keramdorbased on clays from various deposits.

The researcher in (Solov'evetal, 1984) studied clay feedstock in areas with scarce reserves of non-rock materials aiming at determination of its suitability for production of high strength ceramicfiller. On the basis of the obtained results, a new type of high strength ceramicmaterial was developed based on clay feedstock known as keramlit gravel, characterized by active shell on its grains with the thickness of 0.1-0.15 mm comprised of mix of carbonate rock (limestone) with clay.

At present, the investigations devoted to the use of bulk wastes for production of light fillers become more and more important (Abramovetal, 2013; Sharonovaetal, 2006).

The new developed technology is based on approaches to processing of fine wastes of coal beneficiation for production of fly ash aggregate. Fly ash aggregate as porousfiller is efficient in construction light concretes and in road construction.

In comparison with lightweight expanded clay aggregate, the fly ash aggregate produced by the developed technology is characterized by increased strength and low average density. Moreover, final products are characterized by low price due to production on the basis of wastes as the main feedstock.

In (Dengetal, 2018), ash is used as the main feedstock for production of ceramicbricks. The produced ceramic bricks are characterized by improved physicochemical properties and do not evolve harmful environmental pollutants.

The field of ash application becomes wider and wider. As a consequence of combined sintering of ash and clay, it is possible to obtain micro-filtering membrane for purification of oil-in-water emulsion. Mullite fiber crystals provide

excellent heat resistance and steady shrinkage, improving porosity and tensile strength of substrate. As a consequence, the feedstock price, energy consumptions, and duration of production of ceramic membranes are decreased (Zouaetal, 2019).

Ash, together with other industrial wastes was used for production of partially glassy porous ceramicmaterial. The experimental resultshave demonstrated that increase in sintering temperature can accelerate formation of liquid phase and increase pore sizes, which provides higher porosity and lower bulk density. With ash addition, the porous structure is characterized by uniform size and distribution. Therefore, it is possible to achieve good distribution, high porosity, and high compression strength. Moreover, partial glass formation of ceramic foamed materials improves their chemical resistance (Taoyong Liuaetal, 2019).

One of promising trends of ash application is production of light fillers in combination with glass wastes. Powdered glass produced by milling of window glass wastes are mixed with ash and sintered to produce light fillers. After sintering of the mix at 1,050°Cand 1,250°C, new crystalline phases are produced, such as diopside ($MgCaSiO_6$) and wollastonite ($CaSiO_3$), which improves sintering due to relatively low fusing points and porous crystalline structure of light filler(Yu-Ling Weietal, 2016).

One of strategic trends of modern industrial construction in Kazakhstan is improvement of energy efficiency of buildings and structures by decrease in weight of precast concrete and reinforced concrete structures and their heat conductance.

In this regard, substitution of natural gravel with light artificial porous fillers is highly efficient decreasing the structure weight by 20-50%. Herewith, heat loss of building decreases, the level of their heat protection and steam permeability improves, thus improving the standard of living (Yarmakovskiietal, 2012).

Therefore, development of materials of various purposes with high level of operational properties is the main task of modern materials science. Such approach makes it possible to implement cardinally novel engineering approaches to production of efficient materials.

This work is aimed atstudying compositions based on clay rocks and ash of power stations for production of sinteredmicro-porousgranulatedceramicfiller. The formulated target was based on solution of the following tasks:

- Selection of research methods and feedstocks for production of sinteredmicro-porousgranulated
- ceramicfiller;
- Studying chemical mineralogical and physicochemical properties of the selected feedstocks;
- Development of ceramic compositions based on clay–ash system and
- Studying major variations of physicochemical properties of sinteredmicro-porousgranulatedceramic
- filleras a function of sintering temperature and composition.

MATERIALS AND METHODS

The main feedstock was the clay of West Kazakhstan deposit, and the modifying agent was the ash of Ekibastuz Power Station. Clay was sampled directly at the deposit, and the ash of Ekibastuz Power Stationwas taken from waste dumps.

Clays were sampled and their properties in laboratory were determined according to the appropriate regulations: State standard GOST 5180, State standard GOST 12248, State standard GOST 12536, State standard GOST 22733, State standard GOST 23161, State standard GOST 23740, State standard GOST 24143, State standard GOST 26263, and State standard GOST 30416.

Ash of Ekibastuz Power Station was sampled, and its properties in laboratory were determined according to the appropriate regulations: State standard GOST 25818-91, Fly ash of thermal power station for concretes. Specifications:

The delivered samples of clay and ash were analyzed in order to determine their physicochemical properties and chemical mineralogical composition aiming at revealing their suitability for production of sinteredmicro-porousgranulatedceramicfiller.

X-ray phase analysis was performed using DRON-3 diffractometer with CuKα radiation in the angle range of 8°–64°. Sensitivity of the method was from 1 to 2%. X-ray phase analysis was performed with powdered clays passing through 0.315sieves.

Chemical mineralogical composition of the considered components was determined using a JSM-6390LV scanning electron microscope equipped with energy dispersion analyzer, an X'Pert PRO MPD X-ray diffractometer, an ICP-MS Agilent 7500cx inductively coupled plasma mass spectrometer (JEOL, Japan)

The West Kazakhstan clay is characterized as follows:

- coefficient of sensitivity to Chizhsky drying: 63–66;
- plasticity number: 10–11;
- average density, kg/m^3: 1,210-1,240;
- chemical mineralogical composition of West Kazakhstan clay is characterized by the absence of montmorillonite. There are mixed layered formations with hydrous mica and kaolinite.

The following crystalline phases exist in the clay: quartz, d/n = 4.23; 3.34; 1,974; 1.813; 1.538*10$^{-10\ M}$, feldspar, d/n = 3.18; 2.286*10$^{-10\ M}$, calcite, d/n = 3.02; 2.018; 1.912*10$^{-10\ M}$, and hematite, d/n = 1.839; 1.686; 1.590*10$^{-10\ M}$.

Figure 1 illustrates the experimental results of West Kazakhstan clay obtained using scanning electron microscope (SEM).

Table 1

Element	Wt%	At%
C	7.99	12.92
O	50.94	61.83
Na	0.64	0.54
Mg	1.41	1.13
Al	5.47	3.94
Si	19.40	13.42
K	1.66	0.82
Ca	7.58	3.67
Ti	0.28	0.11
Mn	0.08	0.03
Fe	4.54	1.58

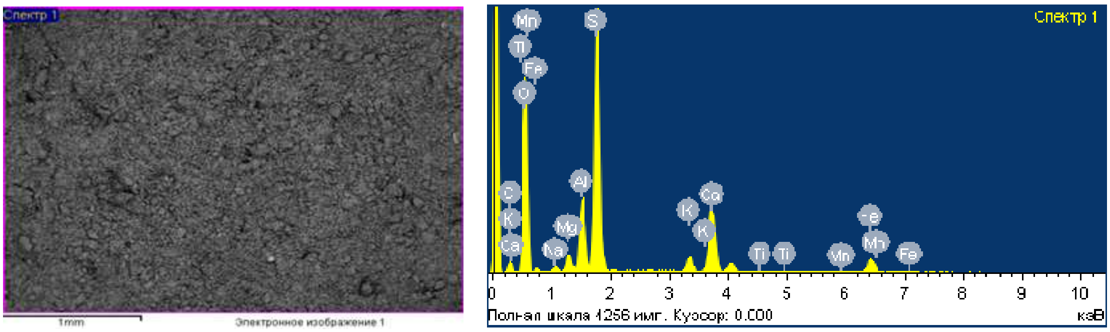
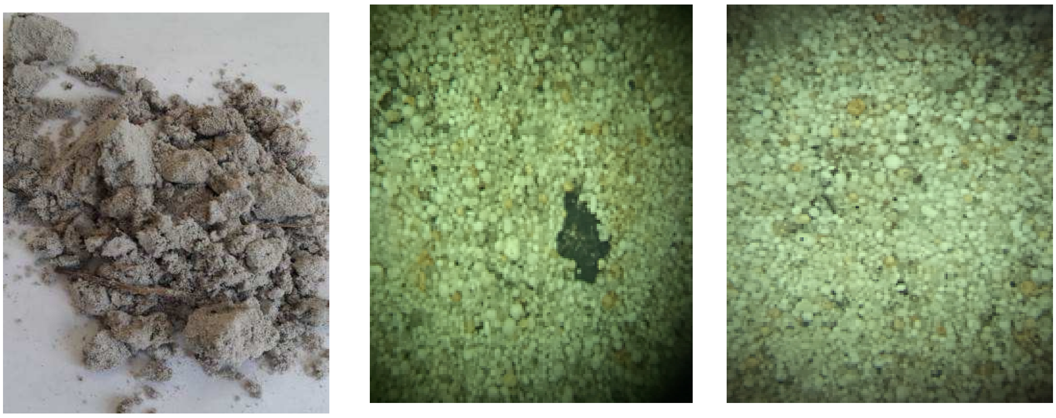


Figure 1: Analysis of West Kazakhstan Clay by Scanning Electron Microscope (SEM)

The other considered substance, the ash of Ekibastuz Power Station, in initial form is loose powdered material of gray color. Under microscope, the ash is comprised of microspheres. Un-burnt coal particles can be seen between the microspheres (Figure 2).



a) Natural Form **b) Macrostructure By Microscope (50×)**

Figure 2: Morphology of Ash of Ekibastuz Power Station

The ash particle size distribution, mm: more than 0.25: 5.98%; 0.25-0.05: 34.8%; 0.05-0.01: 43.07%; 0.01-0.005: 6.55%; 0.005-0.001: 6.40%; less than 0.001: 4.35%.

Specific surface area, cm^2/g: 3200-3700;

Real density, g/cm^3: 1.75-1.84;

Bulk density, kg/m^3: 675-740.

Chemical composition of the ash of Ekibastuz Power Stationis summarized in Table 2.

Table 2: Chemical Composition of Ash of Ekibastuz Power Station

Raw stuff	Content of oxides. wt%												
	SiO_2	Al_2O_3	TiO_2	CaO	MgO	Fe_2O_3	P_2O_5	F	SO_3	CO_2	Na_2O	K_2O	LOI
Ash of Ekibastuz Power Station	57.7	24.5	-	1.10	1.0	4.1	-	-	0.13	-	1.57	-	8.70

The following quantitative mineralogical composition of the ash of Ekibastuz Power Station was determined by X-ray phase and SEM analyses:amorphous clay aggregates– 65-70%, glassy phases – –10-15%, organic matters –10-12%, feldspar –5-7%, hydrogarnet, mullite, iron oxides –5-6%.

RESULTS AND DISCUSSIONS

The ceramic compositing was studied in the following range of ultimate concentrations, wt %:

- West Kazakhstan clay: 70-90;
- Ash of Ekibastuz Power Station: 10-30.

Reference samples without additives were used for comparative analysis.

Feedstocks for studying physicochemical properties were prepared as follows:

At first, the feedstocks were dried in electric drier at 70– 80°Cto residual moisture content of 5-7%. The dried clay was milled in laboratory ball mill to the particle size less than 1mm. The ash of Ekibastuz Power Stationdue to its fine dispersity was used without preliminary milling.

The obtained powdered feedstocks were weighed using electronic scales. Weighed feedstocks were placed to metallic spherical cup for subsequent mixing with water.

In the spherical cup the feedstocks were dry mixed in order to obtain homogeneity. In the dried mix, water was added in amount of 20-28%of weight of dry material.

After addition of water, the feedstock was agitated to homogeneous ceramic paste. The obtained ceramic paste was used for production of granules with the following sizes: 5-10 mm, 10-20 mm, 20-40 mm. Then the granules were dried at 70-80°C to constant weight.

The dried granules were annealed in rotary kiln at 1,000°C (Figure 3).

After burning, the granules were tested.

The annealed samples were dense sintered granules of light red color (Figure 4).

Figure 3: Annealing of Granules in RSR120/1000/13 Rotary Kiln (Germany)

a) **b)**

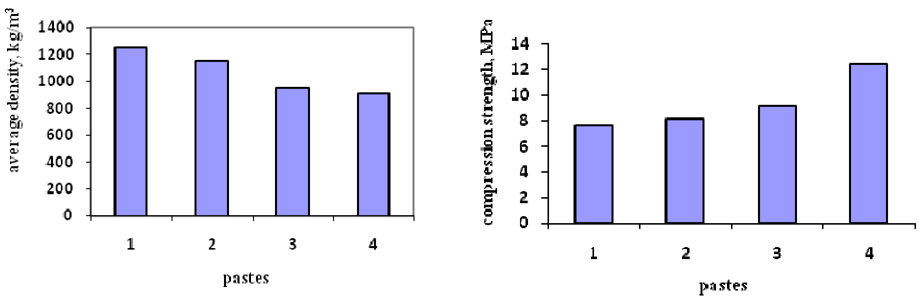
Figure 4: General View of Granules: a) Green Granules, b) Annealed Granules

The following properties of the samples were considered: bulk density, kg/m^3, compression strength in cylinder, MPa, water absorption%, and heat conductance, W/mK. Compositions of the ceramic pastes are summarized in Table 3.

Physico-mechanical properties of the samples are illustrated in Figures 5, 6.

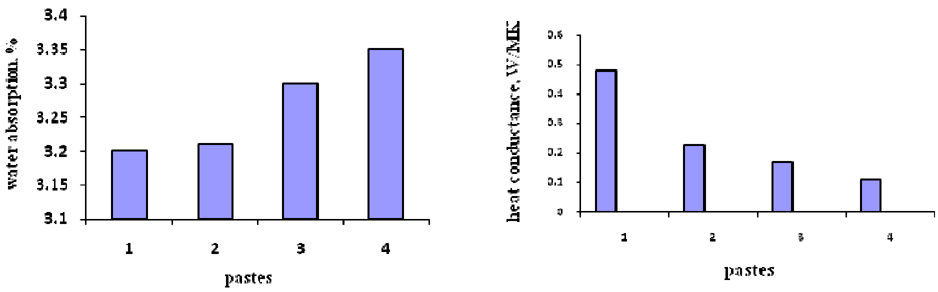
Table 3: Compositions of Ceramic Pastes

Paste No.	Raw compositions
1	Clay - 100%
2	Clay - 90%; ash of Ekibastuz Power Station - 10%
3	Clay– 80%; ash of Ekibastuz Power Station - 20%
4	Clay– 70%; ash of Ekibastuz Power Station – 30%



a) **b)**

Figure 5: Average Density (a) and Compression Strength (b) as a Function of Clay–Ash Compositions



a) **b)**

Figure 6: Water Absorption (a) and Heat Conductance (b) As a Function of Clay–Ash Compositions

The following regularities have been revealed based on variations of physicochemical properties of the considered samples as a function of feedstock compositions: the keramdorsamples based on pure clays can be sintered and form strong structure at 1,000°C. This is evidenced by strength properties at maximum annealing temperature (7.36-7.5 MPa).

With addition of the ash of Ekibastuz Power Stationin amount from 10 to 30%, the strength properties of keramdor samples also increase. Thus, at annealing at 1,000°C, the strength properties increase to 12.4 MPa.

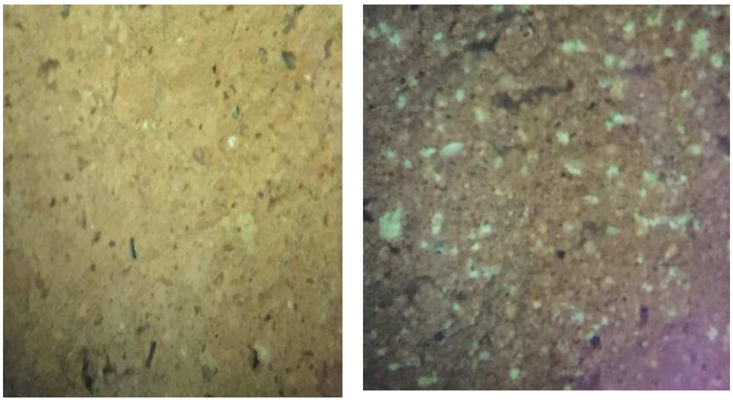
Herewith, average density of the samples decreases and equals to from 1,250kg/m^{3}to 910kg/m^3.

The experimental results also evidence intensification of sintering and crystallization in ceramic pastes containing the ash of Ekibastuz Power Station.

According to X-ray phase and SEM analyses, it has been established that the crystalline phases are comprised of quartz, mullite needle-like crystals, fused grains of feldspar, and hematite. The content of glass phases increases due to partial melting of clay minerals. Formation of crystalline and glass phases at 1,000°C promotes formation of porous strong microstructure of granules.

Decrease in average density can be attributed to formation of porous structure in samples due to burning of un-burnt coal particles contained in the ash. This is evidenced by analysis of microstructure of granules without ash and of granules with added ash in amount up to 30%.

The samples without ash are characterized by dense sintered structure with insignificant micropores. The samples with ash in amount up to 30%are characterized by developed sintered porous structure (Figure 7).



a) b)

Figure 7: Microstructure of Granules: a) w/o Ash; b) with Ash up to 30%

As a consequence, the sintered porousstructure decreased significantly the coefficient of heat conductance. The decrease was from 0.48 to 0.11W/mK.

CONCLUSIONS

- Present state of studies devoted to production of natural and artificialfillers in the form of ceramic fillers and lightweight expanded clay aggregate has been reviewed.
- It has been established that the main efforts in the field of production of ceramic fillers are aimed at the use of various natural and manmade feedstocks.

- Development of resource and energy saving production of ceramic fillers is more and more widely based on nonconventional feedstocks including various industrial wastes. Hence, it is required to study in more details specific features of the applied feedstocks.
- On the basis of experimental studies of physico-mechanical and chemical mineralogical properties of West Kazakhstan clay rocks, it has been established that they can be used as main feedstock for production of granulated sintered ceramic fillers.
- It has been established that the use of bulk secondary feedstocks, for instance, ash of Ekibastuz Power Station, as modifiers improves physicochemical and engineering properties of sintered ceramic fillers.
- According to X-ray phase and SEM analyses, it has been established that the crystalline phases are comprised of quartz, mullite needle-like crystals, fused grains of feldspar, and hematite. The content of glass phases increases due to partial melting of clay minerals. Formation of crystalline and glass phases at 1,000°Cpromotes formation of porous strong microstructure of granules.
- It has been established that the use of ash of Ekibastuz Power Stationin ceramic paste enhances heat conductance of sintered microporous granulated ceramic filler. The use of such materials improves energy efficiency upon construction of buildings.

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